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Spin screening of magnetic moments in superconductors

F. S. Bergeret [1,2], A. F. Volkov [1,3] and K. B. Efetov [1,4]

[1] *Theoretische Physik III, Ruhr-Universität Bochum - D-44780 Bochum, Germany*

[2] *Laboratorio de Física de Sistemas Pequeños y Nanotecnología, CSIC Serrano 144, E-28006 Madrid, Spain*

[3] *Institute of Radioengineering and Electronics of the Russian Academy of Sciences 103907 Moscow, Russia*

[4] *L. D. Landau Institute for Theoretical Physics - 117940 Moscow, Russia*

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Abstract. – We consider ferromagnetic particles embedded into a superconductor and study the screening of their magnetic moments by the spins of the Cooper pairs in the superconductor. It is shown that a magnetic moment opposite to the one of the ferromagnetic particle is induced in the superconductor. In the case of a small itinerant ferromagnet grain and low temperatures, the full screening of the magnetic moment takes place, *i.e.* the absolute value of the total magnetic moment induced in the superconductor is equal to the one of the ferromagnetic particle. In type-II superconductors the proposed screening by spins of the conduction electrons can be much stronger than the conventional screening by Meissner currents.

The phenomenon of screening is very common in physics. The best-known example is the screening of an electric charge in metals due to a redistribution of free electrons in space. This charge screening is very strong and the length characterizing an exponential decay of the electric field (the Thomas-Fermi length λ_{TF}) is in most metals of the order of the Fermi wavelength, *i.e.* of the order of the interatomic spacing. The length λ_{TF} does not depend on whether the metal is in the normal or in the superconducting state.

Another famous example is the screening of a magnetic field or a magnetic moment by superconducting currents in a superconductor (Meissner effect) [1]. Due to this effect the magnetic field decays over the length λ_L (the penetration depth) and vanishes in a bulk superconductor. The same length characterizes the decay of the magnetic field created by a ferromagnetic (F) grain embedded in a superconductor. In contrast to the screening of the electric charge in normal metals, the screening of the magnetic field and magnetic moments in superconductors is weaker and the penetration depth λ_L can be of the order of hundreds interatomic distances or larger. The screening of the magnetic moment is a phenomenon specific for a superconductor and, in contrast to the charge screening, is very small in a normal metal. Although the stray field created by a ferromagnetic grain embedded in a nonmagnetic (N) metal may induce a negative local magnetization in some regions of the normal metal due to the Pauli paramagnetism, the susceptibility is rather small ($\mu_{\mathrm{B}}^2\nu \sim 10^{-6}$, μ_{B} is the Bohr magneton and ν the density of states at the Fermi level) and the screening can be neglected. For certain geometries of a superconducting sample (films, wires) the penetration length λ_L can exceed the transversal size and the screening of the magnetic moment of a ferromagnetic particle due to the Meissner currents does not play an essential role. It is usually believed that in such a situation the total magnetic moment is just the magnetic moment of the ferromagnetic particle and no additional magnetization is induced by the electrons of the superconductor. This common wisdom is quite natural because, at first glance, a possible contribution into the screening of the electron spins in the superconductor is even smaller than in the normal metal. In conventional superconductors the total spin of a Cooper pair is equal to zero and the polarization of the conduction electrons is even smaller than in the normal metal. Spin-orbit interactions may lead to a finite magnetic susceptibility of the superconductor [2], but it is positive and anyway smaller than in the normal state.

In this letter we suggest a new mechanism for the screening of the magnetic moment of a ferromagnetic particle embedded in a superconductor by spins of the superconducting electrons. This effect is large and the magnetic moment of the grain can be completely screened. The characteristic length of the screening is of the order of the size of the Cooper pair $\xi_{\mathrm{S}} = \sqrt{D_{\mathrm{S}}/2\pi T_{\mathrm{c}}}$ (we consider the "dirty" limit) and can be much smaller than the penetration depth λ_L in type-II superconductors. If the size of the superconductor in the transverse direction is smaller than the penetration length λ_L, the mechanism we propose is the only one leading to the screening of the magnetic moment of the ferromagnetic grain.

Although this effect seems very surprising and has been overlooked in all previous investigations of superconductivity, its origin can be understood without any calculations. This additional screening arises due to the exchange interaction between the spins of the conduction electrons and the magnetic moment of the F grain, and the possibility for the superconducting condensate to penetrate the grain.

If the size of the grain is much smaller than the size of the Cooper pair ξ_{S}, the probability that both electrons of a Cooper pair are located in the ferromagnetic grain is small. Therefore, one can assume that only one electron of the Cooper pair spends some time in the grain. Then, the exchange interaction enforces the spin of this electron to be parallel to the magnetization in the grain. This leaves no choice for the second electron in the Cooper pair but to be antiparallel to the magnetization of the grain. In this way, an additional magnetization antiparallel to the one in the grain is induced in the superconductor. Of course, if the transparency of the S/F interface is small, the induced magnetization is weak. However, if the transparency is high enough, the induced magnetization is large and the magnetization of the grain can be completely screened.

Below we support this qualitative discussion by an explicit calculation using quasiclassical Green's functions and a proper Usadel-type equation including the exchange field. Although the calculational scheme is quite standard, it contains a new important ingredient: the superconducting condensate in the F region near the surface consists of a singlet and a triplet components [3]. Due to the proximity effect, the triplet component penetrates the superconductor over the length ξ_{S} and, as it carries spin, polarizes the superconductor. The triplet component was not considered previously for this type of problems. We consider a ferromagnetic spherical grain of radius a embedded in a superconductor S (fig. 1). The superconductor is assumed to be a conventional s-wave superconductor with singlet pairing. The Hamiltonian describing the system can be written as

$$H = H_0 - \sum_{\{p,s\}} \left\{ a^{\dagger}_{ps} J \hat{\sigma}_3 a_{ps'} + \left(\Delta a^{\dagger}_{\bar{p}\bar{s}} a^{\dagger}_{ps} + \text{c.c.} \right) \right\}, \tag{1}$$

where H_0 is the one-particle electron energy including interaction with impurities, J is the

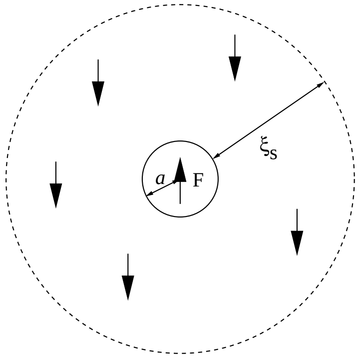

Fig. 1 – Schematic representation of a magnetic grain embedded in a superconductor. The arrows show the direction of magnetization. In the superconductor a negative magnetization is induced over distances of the order of ξ_{S}.

exchange field which is nonzero only inside the F grain, and Δ is the superconducting order parameter which vanishes inside F. The matrix $\hat{\sigma}_3$ is the Pauli matrix in the spin-space and we have assumed that the magnetization of the grain is in the z-direction. The notation $\bar{p}$ and $\bar{s}$ means inversion of momentum and spin, respectively.

In order to calculate physical quantities of interest, it is convenient to use quasiclassical Green's functions $\check{g}$ and write the proper quasiclassical equations for them. We consider the "dirty" limit, which means that the inverse momentum relaxation time τ^{-1} is much larger than the exchange energy J and than the critical temperature T_{c} of the superconductor. The functions $\check{g}$ are 4×4 matrices in the particle-hole and spin space and obey the Usadel equation which, in the presence of the exchange interaction, has the form

$$D\nabla\big(\check{g}\nabla\check{g}\big) - \omega\big[\hat{\tau}_3\hat{\sigma}_0, \check{g}\big] + iJ\big[\hat{\tau}_3\hat{\sigma}_3, \check{g}\big] = -i\big[\check{\Delta}, \check{g}\big]. \tag{2}$$

In the S region $D = D_{\mathrm{S}}$, $J = 0$, $\check{\Delta} = \Delta i\hat{\tau}_2\hat{\sigma}_3$ (the phase of Δ is chosen to be zero). In the F grain $D = D_{\mathrm{F}}$ and $\Delta = 0$. Equation (2) should be complemented by boundary conditions at the S/F interface [4]

$$\gamma_{\mathrm{F}}\big(\check{g}\boldsymbol{n}\nabla\check{g}\big)_{\mathrm{F}} = \gamma_{\mathrm{S}}\big(\check{g}\boldsymbol{n}\nabla\check{g}\big)_{\mathrm{S}}; \qquad \gamma_{\mathrm{F}}\big(\check{g}\boldsymbol{n}\nabla\check{g}\big)_{\mathrm{F}} = -\big[\check{g}_{\mathrm{S}}, \check{g}_{\mathrm{F}}\big], \tag{3}$$

where $\gamma_{\mathrm{S,F}} = \sigma_{\mathrm{S,F}}R_b$, $\sigma_{\mathrm{S,F}}$ are the conductivities of the F and S layers, R_b is the resistance per unit area, and $\boldsymbol{n}$ is the unit vector normal to the S/F interface. Solving eqs. (2), (3) for the Green functions $\check{g}$, one can obtain the induced magnetization δM:

$$\delta M = \mu\delta\sum_p \Big(\Big\langle c^{\dagger}_{p\uparrow}c_{p\uparrow} - c^{\dagger}_{p\downarrow}c_{p\downarrow}\Big\rangle\Big) = -\mu i\pi\nu T\sum_{\omega=-\infty}^{\omega=+\infty}\mathrm{Tr}(\hat{\sigma}_3\hat{g})/2\,, \tag{4}$$

where μ is an effective Bohr magneton, ν is the density of states (DOS) and the sum is taken over the Matsubara frequencies $\omega = \pi T(2n+1)$.

The functions $\check{g}$ can be represented in the form $\check{g} = i\hat{\tau}_2\hat{f} + \hat{\tau}_3\hat{g}$, where the condensate function $\hat{f}$ and the normal function $\hat{g}$ are matrices in the spin space [3]. For simplicity, we consider the case of a small grain when the condition $a \le \xi_{\mathrm{F}} = \sqrt{D_{\mathrm{F}}/J}$ is fulfilled (this is possible provided the exchange energy J is much smaller than the Fermi energy ε_{F}). In this limit the solution can be found by averaging eq. (2) over the grain volume and we obtain for the Green functions

$$g_{\mathrm{F}\pm} = \tilde{\omega}_{\pm}/\zeta_{\omega\pm}\,, \qquad f_{\mathrm{F}\pm} = \pm\epsilon_{b\mathrm{F}} f_{\mathrm{BCS}}/\zeta_{\omega\pm}\,. \tag{5}$$

Here $g_{\mathrm{F}\pm}$ and $f_{\mathrm{F}\pm}$ are the diagonal elements of the matrices $\hat{g}$ and $\hat{f}$, $\tilde{\omega}_{\pm} = \omega + \epsilon_{b\mathrm{F}} g_{\mathrm{BCS}} \mp iJ$, $\zeta_{\omega\pm} = \sqrt{\tilde{\omega}_{\pm}^2 - (\epsilon_{b\mathrm{F}} f_{\mathrm{BCS}})^2}$, $g_{\mathrm{BCS}} = i(\omega/\Delta) f_{\mathrm{BCS}} = \omega/\sqrt{\omega^2+\Delta^2}$, $\epsilon_{b\mathrm{F}} = 3D_{\mathrm{F}}/(2\gamma_{\mathrm{F}} a)$. When obtaining eq. (5), we assumed that the functions $\hat{g}_{\mathrm{S}}$, $\hat{f}_{\mathrm{S}}$ are close to their BCS values $\hat{g}_{\mathrm{BCS}}$, $\hat{f}_{\mathrm{BCS}}$, *i.e.* the corrections $\delta\hat{g}_{\mathrm{S}}$, $\delta\hat{f}_{\mathrm{S}}$ are small. Under this assumption eq. (2) for the functions $\delta\hat{g}_{\mathrm{S}}$, δf_{S} can be linearized. For example, the linearized Usadel equation for $\delta g_{\mathrm{S}3} \equiv \mathrm{Tr}(\hat{\sigma}_3\hat{g})/2$ has the form

$$\nabla^2 \delta g_{\mathrm{S}3} - \kappa_{\mathrm{S}}^2 \delta g_{\mathrm{S}3} = 0. \tag{6}$$

The function $\delta g_{\mathrm{S}3}$ determines the excess magnetization. Solving eq. (6) with the boundary condition, eq. (3), we obtain

$$\delta g_{\mathrm{S}3} = \frac{f_{\mathrm{BCS}}}{\gamma_{\mathrm{S}}} (g_{\mathrm{BCS}} f_{\mathrm{F}0} - f_{\mathrm{BCS}} g_{\mathrm{F}3}) \frac{a^2}{1+\kappa_{\mathrm{S}} a} \frac{e^{-\kappa_{\mathrm{S}}(r-a)}}{r}\,, \tag{7}$$

where $\kappa_{\mathrm{S}}^2 = 2\sqrt{\omega^2+\Delta^2}/D_{\mathrm{S}}$, $f_{\mathrm{F}0} = (f_{\mathrm{F}+} - f_{\mathrm{F}-})/2$ describes the triplet component mentioned above. One can see that the correction $\delta g_{\mathrm{S}3}$ is small if the parameter a/γ_{S} is small. Note also that the function $\delta g_{\mathrm{S}3}$, which, according to eq. (4) determines the induced magnetization, is proportional to the condensate function $f_{\mathrm{F}0}$. The latter function is the triplet component with zero projection of the magnetic moment on the z-axis. Using eq. (7), one can easily calculate the total magnetic moment of the S region:

$$\mathcal{M}_{\mathrm{S}} = -i\pi\nu_{\mathrm{S}} T\mu \sum_{\omega=-\infty}^{\omega=+\infty} \int \mathrm{d}^3 r\, \delta g_{\mathrm{S}3}\,. \tag{8}$$

It is not difficult to see that the magnetic moment M_{S} has the sign opposite to J (the magnetic moment of the ferromagnetic particle $M_{\mathrm{F}0}$ is proportional to J), which means that the spins of the conduction electrons screen (at least partially) the magnetic moment $M_{\mathrm{F}0}$ of the ferromagnetic grain.

Let us compare the total magnetic moment induced in the superconductor, eq. (8), with the magnetic moment of the ferromagnetic grain in the normal state $\mathcal{M}_{\mathrm{F}0} = (4\pi a^3/3) M_{\mathrm{F}0}$. The sum in eq. (8) can be computed numerically in a general case. For simplicity, we consider here a limiting case that can be realized experimentally.

We assume that the transmission coefficient through the S/F interface is not small and the condition $\Delta \ll J \le (D_{\mathrm{F}}/a^2)$ is fulfilled. In this case, the expression for $f_{\mathrm{F}0}$ is drastically simplified. To estimate the energy D_{F}/a^2 we assume that the mean free path is of the order of a. For $a = 30\,\text{Å}$ and $v_{\mathrm{F}} = 10^8\,\mathrm{cm/s}$ we get $D_{\mathrm{F}}/a^2 \approx 1000\,\mathrm{K}$. This condition is fulfilled for ferromagnets with the exchange energy of the order of several hundred K. In order to relate $M_{\mathrm{F}0}$ to J, one has to make a certain assumption about the nature of the ferromagnet. If the magnetic moment $M_{\mathrm{F}0}$ is induced mainly by free electrons (an itinerant ferromagnet), one gets $M_{\mathrm{F}0} = \mu\nu_{\mathrm{F}} J$. Then we obtain for low temperatures

$$\mathcal{M}_{\mathrm{S}}/\mathcal{M}_{\mathrm{F}0} = -1. \tag{9}$$

Equation (9) describes a remarkable phenomenon: at sufficiently low temperatures and in the limit of a small grain ($a \le \xi_{\mathrm{F}}$) the magnetic moment of the latter is screened over distances of the order of ξ_{S}. This screening is complete if the magnetization of the F particle is due to the free electrons (itinerant ferromagnet). It can be easily shown that at arbitrary temperatures this ratio is equal to $-(1-n_{\rm n}(T)/n_{\rm e}$, where $n_{\rm e}$ and $n_{\rm n}(T)$ are the density of the total number of electrons and "normal" electrons defined in chapt. 16 of ref. [1].

The compensation of the magnetization of an itinerant ferromagnet by the Cooper pairs is to some extent consistent with the result obtained by Rusinov and Gor'kov some decades ago [5]. They studied the properties of a superconductor with paramagnetic impurities that were assumed to be ferromagnetically ordered. The free electrons interact with the magnetic impurities via the exchange interaction. Their approach (averaging over impurity positions) reduces the problem to finding the magnetization of a superconductor with an effective exchange interaction uniformly distributed in space. It was demonstrated in ref. [5] that the total itinerant magnetization of the system was zero in the limit of low temperatures and not too large exchange energy.

Clearly, the screening obtained here is due to the appearance of the triplet component. So, if the latter is suppressed by other mechanisms, the effect will be reduced. The spin-orbit interaction (SOI) and orbital effects (Meissner currents) are such mechanisms. The other way of thinking of this reduction is that the spins of the electrons of the Cooper pairs are not necessarily antiparallel in the presence of, *e.g.*, SOI and are not as efficient in inducing the magnetization.

In ref. [3] we studied the effect of the SOI on the triplet component penetration into the ferromagnet. Here we can simply use these results to analyze the effect of the SOI on the penetration of the induced magnetization into the superconductor.

If the SOI is taken into account, an additional term of the form $(i/\tau_{\rm so})[\check{S}\hat{\tau}_3\check{g}\hat{\tau}_3\check{S},\check{g}]$ appears in the Usadel equation, where $\tau_{\rm so}$ is the spin-orbit scattering time, $S=(\sigma_1,\sigma_2,\sigma_3\tau_3)$ (see [6] and [3]). Due to this additional term the quantity $\kappa_{\rm S}^2$ in the linearized Usadel equation is replaced by

$$\kappa_{\rm S}^2=\kappa_{\rm S}^2+\kappa_{\rm so}^2\,, \tag{10}$$

where $\kappa_{\rm so}^2=8/D_{\rm S}\tau_{\rm so}$. Therefore the length of the penetration of $f_{\rm S0}$ and of $M_{\rm S}$ into the S region decreases if $\kappa_{\rm S}^2\sim\xi_{\rm S}^{-2}<\kappa_{\rm so}^2$. The spin-orbit scattering leads to an incomplete screening: the total magnetic moment becomes less than that of the itinerant ferromagnet. In principle, one can measure the spatial distribution of the magnetic moment in the S region (see, *e.g.*, [7]) and get a piece of information about the SOI in the superconductors. With the help of eq. (10), this would be an alternative method to measure the strength of the SOI in superconductors, complementary to the measurement of the Knight shift [8]. (The latter is based on the result of ref. [2] that the Knight shift observed in superconductors is due to the SOI). Let us notice that in the presence of SOI the linearized equation for the function $f_{\rm S3}$ that determines a correction to the energy gap due to the proximity effect remains unchanged.

The orbital effects (the Meissner currents) also change the characteristic length of the penetration of the induced magnetization $M_{\rm S}$. Contrary to the case of bulk conventional superconductors, the Meissner currents in S/F structures arise spontaneously even in the absence of an external magnetic field $H_{\rm ext}$. This happens because an internal magnetic field is induced by the ferromagnet. These spontaneous currents were studied, *e.g.*, in refs. [9,10]. In ref. [9] the spatial dependence of the Meissner current in the F film was calculated, whereas the Meissner currents in both the S and F films were computed in ref. [10]. It is of interest to know not only the spatial dependence and the magnitude of the Meissner currents, but also their effect on the penetration length of the TC and on the induced magnetization. Here we study the influence of the Meissner currents on the penetration length of $M_{\rm S}$. For simplicity, we consider first a planar geometry, which allows us to get a simple solution. For a spherical particle we estimate the effect by order of magnitude.

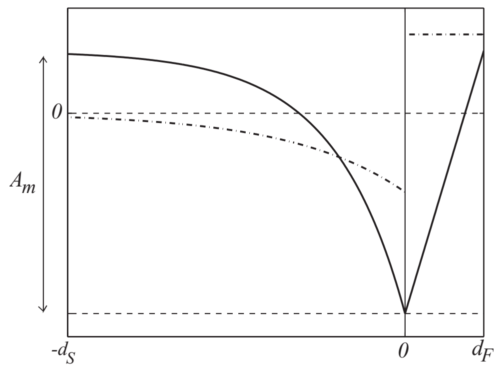

Fig. 2 – Spatial dependence of the vector potential $A(x)$ on the supercurrent density. For low enough temperatures, the values of A at both outer surfaces ($x = -d_{\rm S}$ and $x = d_{\rm F}$) are nearly the same. The dot-dashed line shows the spatial dependence of the local magnetization in arbitrary units.

Let us consider a bilayer S/F structure with the thicknesses $d_{\rm S,F}$. We assume again that the thickness of the F layer is small ($d_{\rm F} \ll \xi_{\rm F}$) and the thickness of the S layer obeys the condition: $\xi_{\rm S} \ll d_{\rm S} \ll \lambda_L$. In this case the solution for $g_{\rm F\pm}$ and $f_{\rm F\pm}$ (eq. (2)) remains unchanged provided one replaces $a/3$ with $d_{\rm F}$. The solution for $\delta g_{\rm S3}$ has the form $\delta g_{\rm S3} = (\kappa_{\rm S} g_{\rm BCS} f_{\rm BCS}/\gamma_{\rm S}) f_{\rm F0} \exp[-\kappa_{\rm S} x]$ and one can easily calculate the spatial distribution of the magnetization in the system $M_{\rm S,F}(x)$. Note that again in the case of an itinerant ferromagnet and low temperatures the total magnetic moment in S is equal to $(-M_{\rm F0} d_{\rm F})$. We display schematically the spatial dependence of the magnetization in fig. 2 alongside with the spatial dependence of the vector potential $A(x)$ which is given by

$$A(x) = A_0 + 4\pi \int_0^x {\rm d}x' M(x') \qquad (11)$$

with a constant A_0 determined from the condition that the total current through the system is zero (no external magnetic field). We have assumed that the magnetization lies in-plane. The supercurrent density is expressed through A as

$$j(x) = 2\pi T \sigma A(x)/(e\phi_0) \sum_\omega f_3^2 \, , \qquad (12)$$

where ϕ_0 is the magnetic flux quantum. In the limit $J \ll \epsilon_{b{\rm F}}$, the condensate functions f_3^2 are nearly the same in the S and F layers. Taking this into account, and calculating the total current I, we get from the condition $I = 0$ for A_0

$$A_0 = -A_m \left(\sigma_{\rm S} d_{\rm S} + \sigma_{\rm F} d_{\rm F}/2 - \sigma_{\rm S} \frac{2\pi T}{D_{\rm S}} \sum_\omega \frac{f_{\rm BCS}^2}{\kappa_{\rm S}^3} \right) / (\sigma_{\rm S} d_{\rm S} + \sigma_{\rm F} d_{\rm F}), \qquad (13)$$

where $A_m = 4\pi M_{\rm F0} d_{\rm F}$. At low temperatures, the third term is approximately equal to $0.53 \sigma_{\rm S} \sqrt{D_{\rm S}/2\Delta}$. The spatial dependence of the vector potential is shown in fig. 2. Equation (7) for $\delta g_{\rm S3}$ does not change in the presence of the Meissner current, provided one replaces $\kappa_{\rm S}^2$ with $\kappa_{\rm S}^2 + p_0^2$, where $p_0 = A_0/\phi_0$. It is clear that the orbital effects are negligible if $A_0/\phi_0 \cong A_m/\phi_0 \ll \xi_{\rm S}^{-1}$. For example, $A_m/\phi_0 \sim 5 \cdot 10^3\,{\rm cm}^{-1}$ for $4\pi M_{\rm F} \sim 1\,{\rm kOe}$ and $d_{\rm F} \sim 50\,$Å, and therefore $\xi_{\rm S}$ should be smaller than $2\,\mu$m.

It is not difficult to estimate p_0 in the case of a spherical particle. The quantity p_0 in this case reaches the maximum at $r = a$ and is of the order of $(4\pi M_\mathrm{F}/\phi_0)a$. As we have noted above, the magnetic moment in F is compensated by that in the superconductor. This means, in particular, that the total internal magnetic flux in the S/F/S Josephson junction may be zero even if the F layer is a single domain (see experiments of ref. [11]).

In summary, we have shown that the magnetic moment $M_{\mathrm{F}0}$ of a ferromagnetic particle embedded into a superconductor is screened by spins of the Cooper pairs. An induced magnetic moment M_S aligned in the opposite to $M_{\mathrm{F}0}$ direction arises in the superconductor.

The induced magnetic moment in the superconductor can be observed experimentally. In a recent experiment [7] the spatial electron spin polarization was determined by means of muon spin rotation. Such a method may be used in order to determine M_S. Another possible method is the measurement of the Knight shift in superconductors that should be dependent on the magnetization M_S. One can also determine the total magnetic moment of a S/F structure performing magnetic resonant measurements as in ref. [12].

At last, one can determine M_S measuring the total magnetic moment of a superconductor with embedded ferromagnetic particles similarly to the work [13], where an enhancement of the magnetic moment of ferromagnetic particles embedded into a nonmagnetic matrix has been observed. Although the enhancement in the case of a normal-metal matrix awaits its explanation, we suggest to measure the magnetization of the system of ferromagnetic particles embedded in a superconducting matrix. When using a superconducting metal as the matrix, a reduction of the magnetic moment, instead of the enhancement, should be observed below the superconducting temperature T_c.

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